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## 3,540,933 REDOX FUEL CELL

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17 Claims

### ABSTRACT OF THE DISCLOSURE

Anolyte and catholyte of same overall chemical composition for redox cell with simple porous separator therebetween and/or each including at least two metallic redox couple complexes. The composition includes metallic complexes having variable valency states (such as ions, oxides, sulphates, phosphates, nitrates, halogenides or other salts) in electrolyte solutions (such as acidic or ionic). Combinations of many complexes of metallic elements in Groups III, IV, V and VI of the Periodic Table having equilibrium potentials relatively close to each other (not more than 0.8 volt apart) are particularly effective in improving power generation. The portion of the electrolyte to be used for the anolyte is reduced by suitable chemical agents and that for the catholyte is oxidized. Suitable electrolyte solutions are acids, such as strong sulphuric or phosphoric acids. Measured amounts of anolyte and catholyte are mixed to have their chemical reaction provide the heat necessary to evaporate the end products resulting from the chemical conversion occurring in the cell. This provides a highly simple and effective means of eliminating waste products such as water.

### CROSS-REFERENCE TO RELATED APPLICATION

The system described and claimed in copending application for U.S. Pat. Ser. No. 652,489, filed July 11, 1967, is advantageously used in conjunction with the invention in this present application.

### BACKGROUND OF THE INVENTION

The invention relates to an electric current-producing cell having a positive and a negative electrode in separate electrode compartments joined by a permeable membrane or partition. The electrolyte or electrically conducting liquid filling the anode- or negative-electrode-compartment is called anolyte, and that filling the positive-electrode- or cathode-compartment is called catholyte. In a redox type of such cell the electromotive force of the cell is derived from a redox couple in the electrolyte. A redox couple is an ionic species which can exist in two valency states: a reduced or red-form and an oxidized or oxform, which are mutually reversible depending upon external chemical or electrical conditions.

At the anode the redox couple oxidizes to its more electropositive form, giving off electrons to the anode and thus charging it negatively with regard to the electrolyte.

At the cathode another redox couple converts to its reduced or electronegative form, taking up electrons from the electrode and thus rendering it electrically positive with regard to the electrolyte.

The anolyte and the catholyte are brought into mutual electrical and material contact through a permeable or porous membrane or separating wall. The wall limits mixing or interdiffusion of the anolyte and catholyte which would destroy the electromotive force. This completes the electrical path inside the cell and electric current is drawn for power use from the oppositely charged

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electrodes until the anolyte is completely oxidized and the catholyte completely reduced.

Recharging of the cell is accomplished by chemically or electrically reducing the anolyte and oxidizing the catholyte. The oxidation can be achieved chemically with air, oxygen, hydrogen peroxide or other suitable oxidizers. The catholyte can be reduced chemically by hydrogen, hydrazine or organic reducers like methanol, which are usually called fuels. A redox cell, depending for the reduction of its catholyte upon a fuel, is usually called a redox fuel cell or chemically regenerative fuel cell.

An example of such a cell is described in the literature (A. M. Posner; Fuel 34 330 (1955)). It has as catholyte either the bromine-bromide or the chlorine-chloride redox couple, and as anolyte the stannic-stannous, or cupric-cuprous, or sulphate-sulphur dioxide redox couple.

In general the rule is stated (K. R. Williams: An Introduction to Fuel Cells; Elsevier N.Y. 1966; on p. 270) that an anode couple is required with a standard redox potential close to zero and a cathode couple with a standard redox potential close to 1.2 volts.

These recent examples are representative of the current state of the art in which it is believed imperative to utilize dissimilar anolyte and catholyte solutions, each based on a single redox couple and which are not permitted to mix or diffuse with each other. German Pat. 264,026 (1913) by W. Nernst describes a cell with more than one redox couple in a common electrolyte but its practical results were so poor that such an approach is presently rejected. Although it was not previously appreciated prior to this invention, the failure of the Nernst approach might be due to its lack of any form of separator to segregate anolyte and catholyte regions in the electrolyte and also its specification that the equilibrium potentials of its redox couples should be substantially more than one volt apart.

It is an object of the invention to provide a novel and improved electrolyte for a redox fuel cell.

It is another object of the invention to provide an electrolyte which can be suitably regenerated with a wide range of fuels, inorganic as well as organic, or by electro-reduction, without the use of solid noble metal catalysts.

### SUMMARY

This invention uses an electrolyte having the same overall chemical composition for both anolyte and catholyte which are loosely segregated from each other by a simple porous separator and/or including at least two redox couple complexes of metallic elements in Groups III, IV, V, and VI of the Periodic System having equilibrium potentials not more than 0.8 volt apart from each other. One complex oxidizes at the anode and one reduces at the cathode to provide the required charge for current flow through the cell. The anolyte and catholyte are suitably pre-reduced and pre-oxidized before connection in circuit. When the overall chemical composition of the electrolyte in the cell is uniform, a simple porous separator can be used between anode- and cathode-spaces without having seepage, leakage, or diffusion through it upset the overall chemical composition or balance of the electrolyte.

Another advantage is, that measured amounts of catholyte and anolyte can be mixed to cause the chemical reaction of their Red- and Ox-forms to provide heat necessary to evaporate the end-products of the chemical conversion of the fuel used, thus preventing these products from accumulating in the electrolyte and impairing its functions.